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**TITLE:** INFLUENCE OF SOIL BIOPOPULATION ON MIGRATION OF WASTE  
RADIONUCLIDES

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# **Influence of Soil Biopopulation on Migration of Waste Radionuclides**

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## **Introduction**

Radionuclides may be removed from waste solutions by various mechanisms employing precipitation or sorption on a medium such as ion exchange resin or soil. Any soluble, nonsorbed material is potentially mobile through the soil in the area of disposal.

In a classical sense, the removal of an inorganic species from solution by sorption on a solid particle may occur by either specific or nonspecific sorption. As defined by Bohn et al,<sup>(1)</sup> specific sorption involves a physicochemical reaction at the surface of the particle whereas nonspecific sorption involves a physical ion exchange reaction at a surface. Nonspecific sorption is most generally encountered in the environment.

The removal of a species is not a simple process as stated; several parameters such as pH, redox potential, ionic strength and complexing ions have an influence on the ionic charge and thus on the removal reactions.

Previous sorption studies using a South Carolina, Fuquay A horizon soil indicated the importance of an entity in that soil relative to its lack of sorptive properties for plutonium. A second observation in those studies related to a general increase in sorption by the soil downward through the soil profile. The least sorptive was the A horizon soil which had the highest content of organic matter.

With respect to the solution in a waste burial pit, it reflects a composite of different wastes from different waste sources. Some of the wastes resulted from clean up procedures which had involved the use of complexing or chelating agents.

In the recent past, clean up procedures often employed some form of chelate

such as versene, tri-vere or sequesterene. Chelated ions thus became a part of the buried waste. The stability of the chelated ion varies with the ionic species, e.g., for EDTA/iron III, the log of the stability constant K, is reported by Welcher<sup>(2)</sup> to be 25.1 whereas the log K for sodium is reported as 1.66 and for plutonium IV the log K is 27.04.<sup>(3)</sup>

Further, some natural organic chelates are produced by microbiological metabolism, e.g., citric acid and oxalic acid. Neilands<sup>(4)</sup> has reported biologically produced hydroxamic acid type siderophores which bind, or chelate, iron and probably plutonium. Because the chelated ions will remain in solution, the presence of chelates in a shallow land burial pit will enhance the potential for migration of radionuclides from the pit and into the soil solution.

This paper reports the interpretation of some results obtained when a Maxey Flats burial pit radioactive waste solution was reacted with a Tilsit soil. The influence of a biopopulation on the degree of sorption and on the stability of that system was investigated.

#### Materials and Methods

Tilsit series soil from the A horizon at the Maxey Flats, KY waste burial site was reacted in the batch mode with liquid waste from trench 19S (4 to 1, wt/wt) at that site. The soil was fortified with 4% by weight dry organic matter. Sets of the soil were prepared in duplicate; appropriate controls were prepared for each set. Distilled water was substituted for waste in each control. A heated and a nonheated series were prepared; the biopopulation in the heated series was controlled by initially heating the waste and soil. Samples were maintained aerobic in a shaking water bath at 32°C.

Sample bottles were removed for chemical and radiochemical analyses at 0 time (less than 1 hour) and after incubation periods of 3, 9, and 53 days. Soluble radionuclides were determined on the supernate after centrifugation of the reaction

mixture to remove solid material calculated to be greater than 0.05  $\mu$  in nominal diameter.

### Results and Conclusions

Results are presented in Table I for sorption of  $^{238}\text{Pu}$  and for  $^{137}\text{Cs}$  in the bioactive or non-heated system. The degree of sorption of  $^{137}\text{Cs}$  on the Tilsit A-horizon soil was similar to that observed for a wide variety of tillable soils collected from the east, the mid-west and the far west.

For the nonheated system, the concentration of soluble  $^{137}\text{Cs}$  was reduced to essentially zero within one hour of contact with the soil. Cesium-137 was not released to the soluble fraction within the 53 day incubation period. Soluble  $^{137}\text{Cs}$  is cationic and univalent; it sorbs readily to soil and normally does not complex.

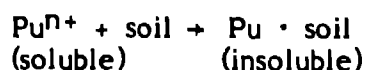
Soluble plutonium also occurs as a cation but in contrast to cesium, plutonium is multivalent and can occur in an ionic form as inorganic and organic complexes. Reduced plutonium, as  $\text{PuIII}$  and  $\text{PuIV}$ , complexes readily with chelates.

In the case of plutonium in the nonheated system, the addition of soil to waste resulted in an increase in soluble  $^{238}\text{Pu}$  of 21% before incubation was initiated. In the previous work cited, employing a Barnwell, SC, Fuquay A-horizon soil, an increase of 200% in soluble  $^{238}\text{Pu}$  had been noted. In both cases, the increases have been attributed to soil entities, e.g. chelates, that solubilize plutonium from the insoluble fraction of the waste. Similar increases have not been observed when lower horizon soils were reacted with plutonium wastes. In the absence of chemical or biological degradation, the soluble plutonium complex will not be sorbed and will migrate with the soil solution. However, if the soluble plutonium complex is degraded by an active biopopulation, the plutonium should be released, should be sorbed by the soil and thus immobilized.

The concentration of soluble plutonium did decrease in the presence of an active biopopulation by 86% during the 53 day incubation period. There was a

concomitant decrease in soluble organic matter (determined as COD) of 73%. The results suggest the presence of a  $^{238}\text{Pu}$ /organic complex that was degraded by the biopopulation; the released  $^{238}\text{Pu}$  was removed from solution by sorption on the soil. The final level of 1.9 pCi/ml remaining in solution at 53 days is considered to reflect a nonbiodegradable chelate under the conditions of the experiment.

The results are expressed as follows:

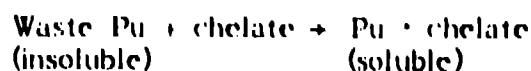


Results from the heated or bioinactive system, are presented in Table II. The rapid sorption of  $^{137}\text{Cs}$  is similar to that observed for the nonheated system. In both cases there was a rapid and high degree of removal of the soluble  $^{137}\text{Cs}$ .

The results indicate that sorptive processes for soluble  $^{137}\text{Cs}$  are active in both systems and that the processes do not relate to an active biopopulation. There is little potential for the migration of  $^{137}\text{Cs}$  from a waste burial pit to the open environment or to an aquifer.

In the case of plutonium, the results obtained for its removal from the heated system were markedly different from those obtained for the nonheated system. For the heated system, the concentration of soluble plutonium remained essentially constant during the 53 day incubation period. Some decrease in the soluble organic matter was noted between 9 days and 53 days. The mean of the respective 4 determinations for  $^{238}\text{Pu}$  is  $1.8 \pm 0.4$  pCi/ml - in fair agreement with the  $1.9 \pm 0.1$  pCi/ml value for the nonheated system at 53 days.

Possible reactions for plutonium in the heated system are shown as follows:



Pu chelate (absence of specific enzymes) → Pu chelate remains in solution (soluble)

The lack of removal of soluble  $^{238}\text{Pu}$  from the heated system suggests complexing by another chelating agent which is heat stable. Results obtained for the nonheated and the heated systems thus suggest the presence of at least two complexing systems. One system is heat labile; the other system is heat stable. The heat labile system is biodegradable within 53 days. The heat stable system, if biodegradable, is metabolized at a slow rate. The complex from that system would be of more concern in the migration of plutonium with the underground flow of soil solution.

#### Summary

The data have been interpreted as follows:

1. The removal of  $^{137}\text{Cs}$  from solution by the soil is essentially complete within a one hour period and is not influenced by an active biopopulation.
2. The soil studied contains complexers which solublize  $^{238}\text{Pu}$ .
3. The soluble complex does not sorb to soil and thus is potentially mobile.
4. In the presence of an active biopopulation 86% of the complexes is degraded; the released  $^{238}\text{Pu}$  was rendered immobile. The remaining 14% of the soluble  $^{238}\text{Pu}$  was not released to the soil during 53 days incubation. That fraction is heat stable and non- or slowly-biodegradable and thus retains its potential to migrate.

#### Acknowledgment

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4. J.E. Neilands, "Microbial Iron Compounds." *Ann. Rev. Biochem.*, 50, 715-31 (1981).

Table I

# Removal of Soluble $^{238}\text{Pu}$ and $^{137}\text{Cs}$ (Bioactive System)

System	Incubation (days)	COD (ppm)	$^{238}\text{Pu}$ (pCi/ml)	$^{137}\text{Cs}$
Waste-total	-	720	42	17
Waste-soluble	-	710	11	17
Waste/As Soil	0	1720	13.4	0.09
	3	1360	9.2	0.05
	9	480	3.7	0.03
	53	470	1.9	0.09



Table II

# Removal of Soluble $^{238}\text{Pu}$ and $^{137}\text{Cs}$ (Bioinactive System)

System	Incubation (days)	COD (ppm)	$^{238}\text{Pu}$ (pCi/ml)	$^{137}\text{Cs}$
Waste-total	—	690	45	1.8
Waste-soluble	—	600	0.5	1.7
Waste/Ap Soil	0	1300	1.6	0.13
	3	1490	1.4	0.00
	9	1660	2.0	0.01
	53	990	2.3	0.05